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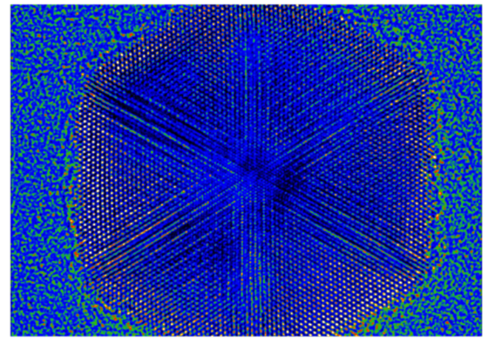
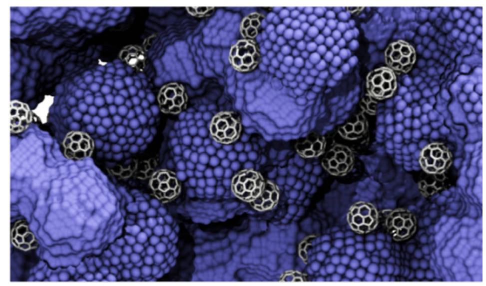
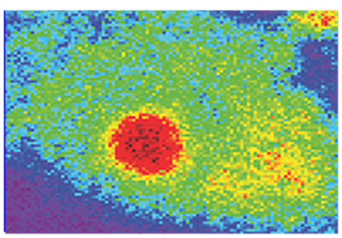
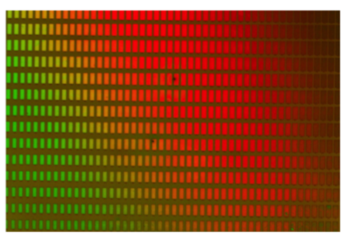
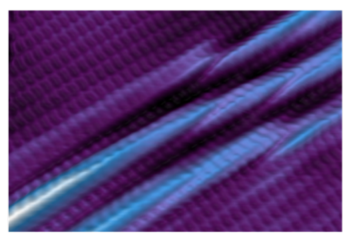
CENTER FOR NANOSCALE MATERIALS

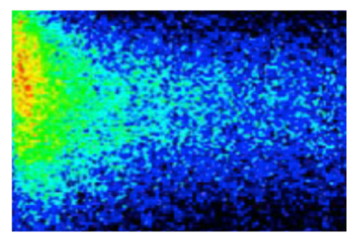












STRATEGIC PLAN 2016-2020

Argonne National Laboratory
Nanoscience and Technology Division
U.S. Department of Energy, Office of Science,
Office of Basic Energy Sciences

U.S. DEPARTMENT OF ENERGY | Office of Science

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Executive Summary

The Center for Nanoscale Materials (CNM) at Argonne National Laboratory is a premier user facility providing expertise, instrumentation, and infrastructure for interdisciplinary nanoscience and nanotechnology research. As a Department of Energy (DOE) funded research center, the CNM is at the forefront of discovery science that addresses national grand challenges encompassing the topics of energy, information, materials and the environment. The Center is also a vibrant member of the Argonne National Laboratory's scientific community, fully invested in the Laboratory's key initiatives for advanced materials and chemistry, and research partnerships with other DOE user facilities at Argonne such as the Advanced Photon Source (APS) and the Argonne Leadership Computing Facility (ALCF).

The scientific strategy of the Center for Nanoscale Materials is consolidated under the following **three crosscutting and interdependent scientific themes**, noted below. Collectively, they aim at the discovery and hierarchical integration of materials across different length scales, at the extremes of temporal, spatial, and energy resolutions:

I) Quantum materials and phenomena: The goal of this theme is to combine CNM's expertise in synthesis, fabrication, characterization and theory on nanometer length scales to discover *fundamental mechanisms and materials for quantum sensing and information*.

II) Manipulating nanoscale interactions: Our goal here is to study and manipulate the forces, the interactions, and the energy dissipation between nanoscale and atomic constituents at interaction lengths that vary from distant (~10 nm), to the atomic scale.

III) Synthesis of nano-architectures for energy, information and functionality: This theme aims to combine synthesis and nanofabrication across different scales to achieve energy efficiency, novel methods of energy transduction and new functional behavior in materials.

Embedded within these three themes, and supporting them are the **vector capabilities of X-ray microscopy, electron microscopy, and computational materials science**. Theme (I) through (III) include requiring detailed atomic understanding of temporal and spatial structural response to applied stimuli, a central theme in the Electron and X-ray Microscopy effort. Computational materials science activity is assuming a leadership position in combing first principles physics and machine learning for new materials discovery related to themes I thru III.

The CNM provides unique capabilities, expertise and tools to its users that include optical spectroscopy from the ultraviolet to the THz at the extremes of spatial and time resolution, the synchrotron X-ray scanning tunneling microscope (SX-STM), the hard X-ray Nanoprobe (HXN), a full suite of STM capabilities, cleanroom-based, comprehensive nanofabrication capabilities, and the Carbon supercomputing cluster. To this the CNM is revamping efforts in electron microscopy that will be aimed at combining data science and electron microscopy. The CNM currently employs 52 staff who contribute to the scientific programs in addition to supporting the users of the facility. During FY16, the CNM hosted 566 users from academia, national laboratories and industry. CNM users, staff, and post-docs are engaged in high-quality science, as evidenced by the publication of a total of 904 journal articles during FY14-16 with 28% of the papers in the top 20 highest impact nanoscience journals as defined by DOE.

In this strategic plan we provide a concise outline of our scientific vision, opportunities, activities and organizational structure. This document will guide our path over the next five years, with the aim of preserving CNM as a world-class user facility where we are ready to provide capabilities users will need 2-5 years into the future, and where our scientists can help shape the future of research in the nanosciences.

1. Introduction

The Center for Nanoscale Materials has been in existence since 2006, and is one of five National Science Research Centers (NSRC) user facilities created and funded by the Department of Energy Office of Basic Energy Sciences, for furthering research in the nanosciences. The Center has a world class staff of 31 scientists and technical personnel housed in five research groups, and is supported by 21 administrative and facilities personnel. It is equipped with leading edge instrumentation and capabilities for synthesis, characterization and testing in the area of nanomaterials and the nanosciences. Since its inception, the CNM has cultivated an engaged and productive user community, becoming a resource used by a wide spectrum of researchers from across the U.S., as well as from Europe, South America, and Asia.

The CNM focuses on working with users on cutting-edge problems and new, innovative science rather than routine measurements. Users come to CNM via a proposal submission and selection process (see section 7 for details). In FY16 there were 566 users. The CNM has strong connections to other facilities and divisions (including joint hires and collaborative projects) within Argonne, such as the Advanced Photon Source (APS), Advanced Leadership Computing Facility (ALCF), the Materials Science Division (MSD), Chemical Sciences and Engineering Division (CSE), Energy Systems Division (ES), etc., and users can also benefit from access to the broader technical community that this provides.

Figure 1-1. *Integrated approach to research at the CNM reflecting close links between the CNM user program, staff expertise, and state-of-the-art facilities for nanoscience and nanotechnology research.*

Figure 1-2. *Center for Nanoscale Materials staff members in 2016.*

Within the user facility framework, the CNM offers access to diverse scientific expertise and an interdisciplinary research culture. Examples include:

- A 12,000 square foot cleanroom (with an additional accessible 6000 sq ft. of space that will be available to users soon), and experienced staff in nanofabrication capable of tackling challenging issues in materials and nanostructural design.
- Access to a high-resolution chromatic aberration corrected TEM, one of only three available worldwide, that can probe thicker samples without loss of resolution and the ability to distinguish between different basis atoms in a cation or anion sublattice.
- A Hard X-ray Nanoprobe (HXN) on APS's beamline 26, capable of 25 nm X-ray beam focusing, demonstrated 5 nm resolution for correlated scanning fluorescence and sub picometer lattice displacement strain resolution in 3-D volumes for in-operando measurements using Bragg ptychography. This is the only dedicated X-ray microscopy facility within the DOE NSRC complex.
- Development (in progress) of the world's first SX-STM user beamline with the goal of orbital level imaging of molecules coupled with chemical identification of atoms.
- Commissioning of quantum measurement tools includes optical microscopy with time-correlated single photon counting capability in the visible and near-infrared (to 2 microns) for time-resolved photoluminescence as well as photon antibunching/correlation studies.
- Time-resolved optical measurement capabilities include extremes of space from tens of nm thru bulk, energy range with less than 10 meV resolution from the ultraviolet (260nm) thru mid-IR (10 microns with THz soon) and time resolution of 70 fs.
- Development of pioneering algorithms that combine machine learning and first principles physics for generating force fields for molecular dynamics calculations with unprecedented accuracy and computing time efficiency.

The list above is only a small subset of CNM's capabilities and is presented here as an example of the unique capabilities that are offered. The CNM considers its role to be one that has both an anticipatory component, where we are ready to provide capabilities users will need 2-5 years into the future, as well as a steering component where our world-class scientists, working with our users and the broader scientific community, help influence and shape the direction where nanoscience should be headed.

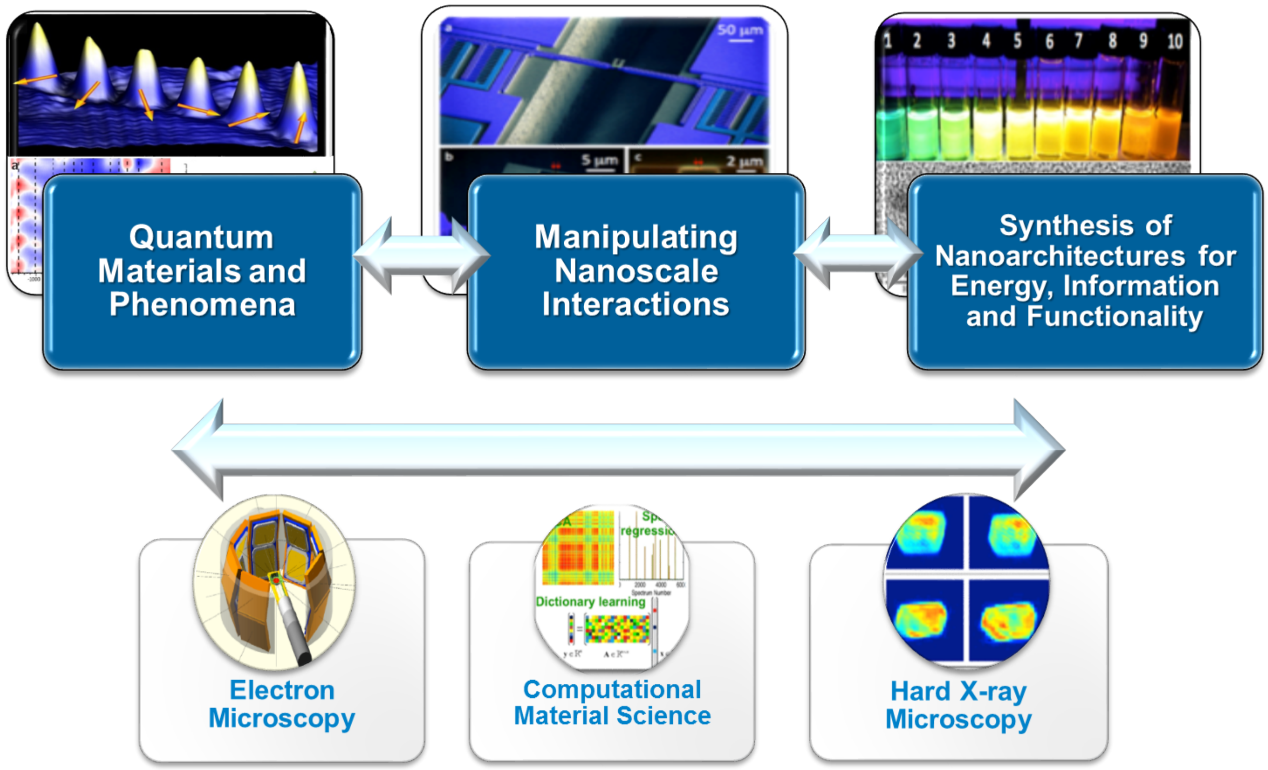


Figure 1-3. *Illustration of CNM's three crosscutting strategic themes, supported by the vector capabilities of electron microscopy, computational materials science and hard X-ray microscopy.*

The scientific strategy of the Center for Nanoscale Materials is consolidated under **three crosscutting and interdependent scientific themes**. This is a highly inter-disciplinary exercise and requires the close, integrated collaboration of five research groups, housing particular specializations, with each group led by a Group Leader. Each theme has a "theme coordinator" who cross-cuts across groups in his or her stewardship of the theme. The three themes are underpinned by three vector capabilities, as shown in Figure 1-3 and further described in Section 2.

2. Research Themes and Groups

2.1 RESEARCH THEMES:

The CNM will pursue the following three themes over the next five years. Our future staffing and "new capability" capital equipment purchases will reflect these directions, as will our over-arching strategy and its planned execution.

I) Quantum materials and phenomena

The goal of this theme is to combine CNM's expertise in synthesis, fabrication, characterization and theory on nanometer length scales to discover *materials and fundamental mechanisms for quantum sensing and imaging, as well as discoveries of novel quantum phenomena in confined structures*.

This theme will include the study of the fundamentals of sub-wavelength light localization, optically and electrically accessible defects, and photon and phonon dynamics in low-dimensional and bulk materials. Specific studies will focus on single photon emission, quantum entanglement, and optical and thermal transport. Development of the materials and understanding of their underlying phenomena within this theme forms a basis for exploring their fundamental interactions studied in Theme (II) and in the nanoarchitectures for energy and information transduction studied in Theme (III).

II) Manipulating nanoscale interactions

The goal of this theme is to build on our *understanding of nanoscale phenomena to achieve active control and manipulation of quantum states, atomistic and nanoscale interactions*, including a focus on dissipation engineering.

A central motif here is to study the forces and the interactions between nanoscale constituents at length scales that vary from the atomic to ~10 nm. These include studying the ability to manipulate and couple nanomechanical elements between themselves and light, the fundamentals of friction at the nanoscale, and the computational simulation of materials and defects from the inter-atomic interactions. Understanding the fundamentals of these nanoscale interactions provides emergent behavior necessary for building nanoscale architectures in Theme (III) and selection of properties in Theme (I).

III) Synthesis of nano-architectures for energy, information and functionality

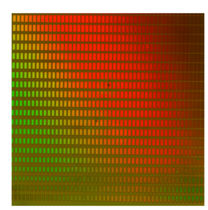
This theme aims to achieve energy efficiency and energy transduction in systems that utilize hierarchical synthesis and materials design at the nanoscale. It combines synthesis and nanofabrication across different scales and aims to use both self-assembly and top-down approaches. The objectives for this approach, in the end, are to identify new pathways in energy and information transduction including propagation of charge, spin and collective phenomena; and develop new ways for adaptive responses to the environment. Implementation of this theme is intimately connected to, and relies on the natural developments arising from themes (I) and (II).

Embedded within these three themes, and supporting them are the **vector capabilities of X-ray microscopy, electron microscopy, and computational materials science**. These will be further discussed in Section 3.

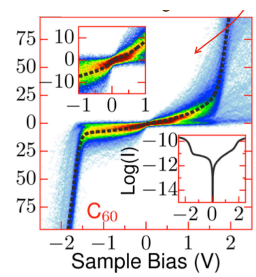
2.2 RESEARCH GROUPS:

The CNM mission for user support and staff science along the three themes identified above is executed through five research groups, divided by specialization and function. These five groups work closely together, as will be discussed below, in support of specific themes. Each of the groups contributes strongly to each of the themes.

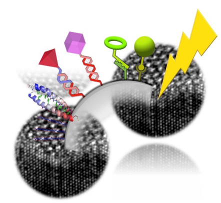
- **The Nanofabrication & Devices Group** (NFD) specializes in the fundamental science behind the development of micro- and nanoscale systems with the goal of achieving unprecedented control in the fabrication, integration, and manipulation of nanostructures. This includes the incorporation – under cleanroom conditions – of materials and active submicron elements that couple mechanical, optical, and electrical signals to produce working nanofabricated structures.



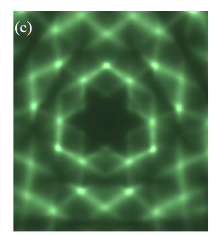
- **The Quantum & Energy Materials Group** (QEM) designs and studies atomic-scale to meso-scale materials with implications for energy, the environment, and coherent information transfer/sensing. Their research includes (i), using a powerful suite of scanning tunneling probe characterization and atomic/molecular manipulation capabilities to "design-in" engineered quantum states down to single atoms, molecules or defects; and (ii), control molecular and nanoscale interactions for the accelerated discovery and fundamental understanding of artificial three- and two-dimensional materials for energy, reaction chemistry, and pollution remediation.



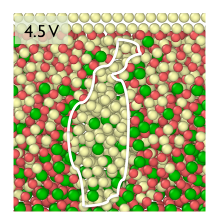
- **The Nanophotonics & Biofunctional Structures Group** (nPBS) studies optical processes at the extremes of time and space resolution through ultra-fast spectroscopies and advanced microscopies. The goal is to understand and realize efficient energy transduction in nanostructures, and study the physics of coherent radiative processes for quantum sensing. The group also seeks to create novel biological assemblies for nature-inspired studies of energy conversion, coherent energy transport, and biosensing mechanisms in cell-like environments functionalized with engineered nanomaterials. The group combines the properties of metals, organics, semiconductors and dielectrics to synthesize efficient catalysts, chemical and biological sensors, and hybrid biological moieties.



- **The Electron and X-ray Microscopy Group** (EXM) develops new research capabilities that go beyond off-the-shelf technology in conjunction with the broadest scientific community that helps identify, define, and develop electron microscopy needs, and advanced X-ray microscopy needs to address the science of the future. Current focus in electron microscopy is to incorporate advances in data science and new modalities for detection into electron microscopy for enhanced interpretation and analysis. The focus in the X-ray microscopy effort is two-fold: (i) use of the synchrotron based hard X-ray Nanoprobe for in-operando pulse/probe studies of dynamic phenomena in materials using Bragg ptychography; and (ii), the development of the synchrotron X-ray scanning tunneling microscope capability.



- **The Theory & Modeling Group** (TMG) works on large scale molecular dynamics, high-level electronic structure theory, quantum and electrodynamics, multi-scale modeling and data science based approaches to understand and predict a wide range of phenomena including nanoscale tribology, thermal and charge transport, and quantum entanglement in hybrid plasmonic systems.



2.3 INTEGRATED APPROACH TO THE THEMES

How are the different groups and activities specifically integrated in their pursuit of the research outlined in the three different themes? Each theme has a theme coordinator who is responsible for oversight over the theme activities, working closely with the scientists from the different groups and the group leaders to ensure the progress of the theme research. Each group is involved in at least 2 themes, and themes also contain participants from other divisions and centers within ANL. In section 3 we provide specific detail for each theme and highlight the integrated and cross-connected approach to CNM's research.

3. Vision for Scientific Growth

3.1 RATIONALE AND METHODOLOGY:

Determining a technical strategy that will maximize our impact to science and our benefit to our users requires us to both anticipate future user needs, as well as help shape future areas of focus in the nanosciences. The development of such a strategy was stewarded by a core team within CNM consisting of the Group Leaders, the Director and the Deputy Director that met weekly for this purpose for a collective period of over 8 months. We followed a deliberative process to developing our technical strategy that took into account a multi-tiered set of inputs, and followed an evolutionary approach:

First, several governmental documents and workshops offered us guidance. These include The White House, DOE and NSF Grand Challenges for the nanosciences. The Basic Energy Sciences Advisory Committee (BESAC) report (November, 2015) from the DOE Office of Basic Energy Science (BES) on *Challenges at the Frontiers of Matter and Energy: Transformative Opportunities for Discovery Science*, with its emphasis on hierarchical assemblies and non-equilibrium structures provided a template for our strategy. Recently concluded BES workshops offered valuable perspective. For instance the recently concluded workshop by the BES *Basic Research Needs for Quantum Materials for Energy Relevant Technology* (February, 2016) highlighted the need for exploring the physics of individual quantum states and entangled states. The workshop titled *Neuromorphic Computing: From Materials to Systems Architecture* highlighted the need for exploring new materials and physics for neuromorphic information processing.

Second, we collected input from a variety of sources. This included formal and informal submission of short ideas/proposals from the staff, statements of accomplishment, committees created for development of specific sub-strategies (such as those in electron and X-ray microscopy), discussions with the User Executive Committee, with key ANL collaborators and stakeholders such as relevant personnel at the Advanced Photon Source (APS), with external members of the academic community, with the members of our Scientific Advisory Committee (SAC), and with key users. The inputs were collected and filtered by the Group Leaders. The strategy was then further shaped via a-day long retreats with participation from the entire CNM technical staff, and presentations and focus group discussions by the staff; and a presentation of our strategy to the SAC committee.

The final strategic plan emphasizes an approach that encompasses our scientific vision as well as provides scientific leadership to bring added value to our users, and in particular attract new user communities.

Our strategy is closely linked to Argonne's strategic plan that guides core research programs at Argonne, namely the advanced materials and chemistry initiative, while synergistically working with other DOE user facilities at Argonne. The CNM is already tightly integrated in its collaboration with the APS, ALCF, ES , MSD, BSD (Biosciences Division) through joint research projects, shared facilities, and jointly authored publications. A part of this interdisciplinary vision setting is made possible via Argonne internal LDRD funding for exploratory projects. In addition to this, CNM is forging relations to the Computing, Environment, and Life Sciences (CELS) Directorate at ANL in order to make the connection to computer science and data science.

3.2. DESCRIPTION OF FUTURE DIRECTIONS:

In the following we describe our specific strategic plans for each of the three themes articulated earlier. We will describe specific projects in order to highlight the overarching goal of each theme: it is not our intent for this to be a comprehensive list of all projects being carried out within CNM. We also indicate how the five different groups interact and engage to accomplish the objectives of the themes. Finally, our equipment purchase plans required to accomplish the work are presented.

3.2.a. Theme I, Quantum materials and phenomena

Quantum materials and phenomena includes studies leading to the creation and manipulation of single quanta of information, and the use of quantum mechanical phenomena to extract maximal information from a sensor probing its environment. This theme will include the study of the fundamentals of sub-wavelength light localization, spins, and optically and electrically accessible defects in low-dimensional and bulk materials for quantum coherence and entanglement. It also includes new opportunities for using quantum phenomena and quantum sensing for making measurements of materials and surfaces—an area particularly significant for user facilities such as the CNM. Finally, it also includes the synthesis of materials and defect geometries required for the studies noted above. Research in this area will focus on:

Optical qubits, sensing, and entanglement

The emerging field of quantum information science holds the key to the next generation energy efficient quantum computing, communication, and quantum metrology techniques. Of the various types of qubits, explored for quantum information, single photons are one of the most promising due to their weak interactions and relative ease for encoding quantum information. Ideal single photon sources for quantum information applications should exhibit high photostability, extraction efficiency and indistinguishability. Defect centers within a semiconductor are an emerging source of single photons—correlating and controlling them is the challenge. We will engineer optically active dopants and defects in nanomaterials and 2D materials (e.g. WSe_2, figure 3-1, left) for high frequency efficient single photon operation. We will then explore coherent interactions between defects by manipulating the spacing and orientation between them using scanning probe techniques. In conjunction with this, photonic quantum information will be encoded so as to realize entanglement between two single photons in photonic circuits. We will also explore the interaction of the single photon sources with metal nanoparticles, i.e. the exciting research frontier of "quantum plasmonics" as a means of controlling the single photon emission characteristics (figure 3-1, right). This may allow further control of correlated photons, leading to highly sensitive single molecule adsorbate sensors. A single photon detection microscope with a spatial resolution up to 150 nm and a temporal resolution of 50 ps has been commissioned for photon autocorrelation measurements in the visible and telecom wavelength ranges for users. This work is a collaboration between the NPB, QEM, and TMG groups.

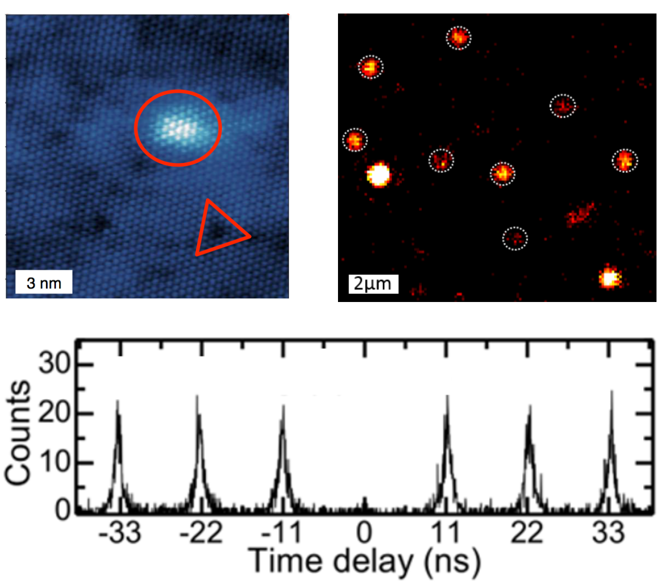


Figure 3-1. *Left: UHV STM image of two different types of defects (marked) in WSe_2. Right: Scanning photoluminescence image of individual dopants in carbon nanotubes (marked by white circles). Bottom: A second-order photon correlation curve of a single dopant in a carbon nanotube. The appearance of photon antibunching indicates the single photon signature of the dopant.*

Dynamics of photons & phonons under quantum confinement

We aim at understanding--experimentally and theoretically—photon and phonon transport and dynamics in nanoscale structures under external stimuli (light, temperature, electric field). As current densities, electron-phonon interactions, and vibrational mismatches all increase in nanoscale materials and at heterogeneous interfaces, we will understand how the "intrinsic" heat generation by electrons can be tuned via chemical synthesis, and how heat transport out of nanoscale architectures can be channeled (Figure 3-2).

A true predictive understanding of the structural control of thermal transport remains limited in part by the lack of appropriate nanoscale scattering tools sensitive to phonon dispersion. This situation has now changed. Using a recently developed (EXM Group) technique of synchrotron Thermal Diffuse Scattering (TDS) as a quantitative tool to observe locally confined phonon dispersion along with a combination of advanced spectroscopic method (Femto-second Raman, transient absorption, nPBS Group), and first-principles calculations of electron-phonon and phonon-phonon interactions (TMG Group), we seek a complete picture of phonon emission and transport across timescales ranging from the femtosecond electron relaxation to the nanosecond heat transport.

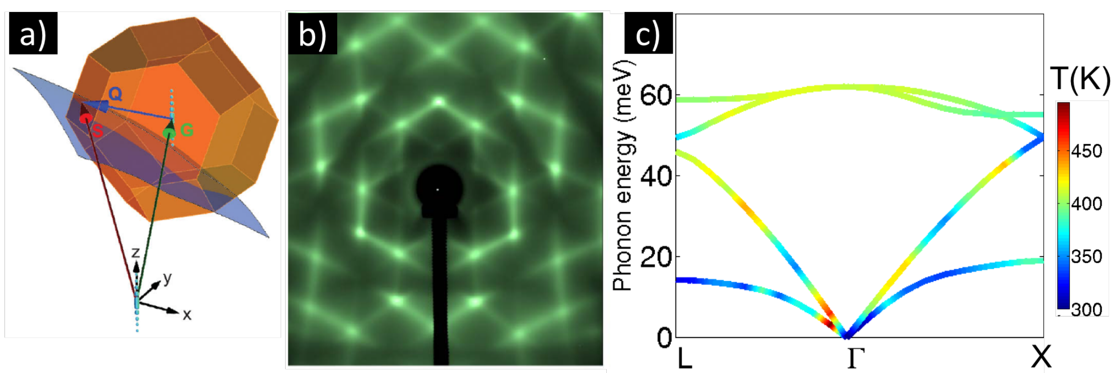


Figure 3-2. *(a) Nanoscale X-ray Thermal Diffuse Scattering (nano-TDS) allows a unique observation of the large wave-vector phonon dispersion and occupations in nanostructured materials [Gopalakrishnan and Holt PRL 2013]. (b) Experimental X-ray TDS image from Si[111] .(c) Computed phonon dispersions and non-equilibrium occupations in silicon 10ps after excitation by hot electrons (T=3000K) using the Boltzmann transport equation interfaced with first-principles calculations of electron-phonon and phonon-ponon interactions.*

Concurrently, we seek to understand the photoinduced dynamics of excitons in quantum confined hybrid nanomaterials for opportunities related to directional optical energy transduction and transport, as well as for development of high quality single photon sources. This is challenging because nanoparticle inhomogeneities can dramatically alter exciton dynamics from particle to particle, thus short circuiting any opportunity for directional energy flow or the opportunity to observe photon entanglement. We plan an active colloidal synthesis effort to create improved core-shell semiconductor nanostructures of high homogeneity and reduced surface traps, including but not limited to novel nanoplatelet materials (Figure 3-3). In collaboration with electrodynamics modeling efforts of the TMG group and metasurface fabrication in the NFD group, this work will extend to solid state environments for coupling to nanostructured metamaterial substrates and photonic circuitry. New directions in size selective nanoparticle assembly will also be pursued, so as to create an energy waterfall effect that produces directional photoinduced exciton flow that can be used for energy transduction. These studies will also extend to novel photoinduced excitations such as biexcitons in nanomaterials for quantum information and quantum entanglement studies.

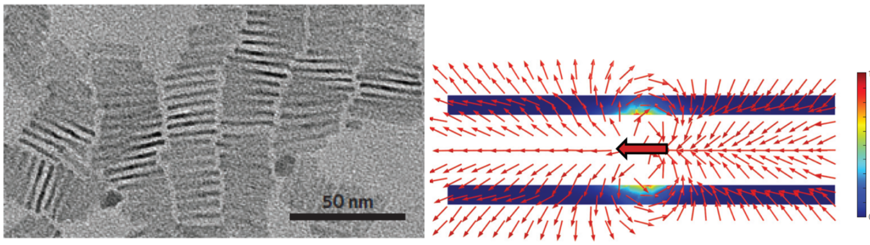


Figure 3-3. *2D assemblies of colloidal CdSe semiconductor nanoplatelets (left) and modeling of energy transfer (right) between platelets are shown (C. E. Rowland et al., Nat. Materials* ***14****, 484 (2015)).*

Orbital and chemical imaging using X-ray excited tunneling spectroscopy

Synchrotron X-ray Scanning Tunneling Microscopy (SX-STM) combines two powerful techniques for materials characterization, scanning tunneling microscope (STM) and X-ray illumination, into a novel approach to nanoscale imaging with elemental, chemical, and magnetic sensitivity. Unlike most synchrotron techniques, where spatial resolution is dependent on the size of the X-ray beam incident on the sample, the spatial resolution in SX-STM is determined by a probe tip located just a few angstroms away from the sample. Therefore, it has a potential to achieve spatial resolution down to the atomic scale. One of our main research goals is to pin-point individual atoms inside single molecules one molecule-at-a-time basis and to investigate their quantum properties up-close and personal using this nascent SX-STM technique. When synchrotron X-ray beam with matching photon energy of an elemental absorption edge is illuminated on the sample, the element specific X-ray excited electrons are generated. SX-STM imaging is based on the tunneling of the X-ray excited electrons that fill up unoccupied molecular orbitals. Thus simultaneous acquisition of elemental, chemical, and magnetic maps of individual atoms inside metallo-organic molecules including proteins becomes a possibility. Metallo-organic molecules are composed of metal ions linked by hydrocarbons and they are technologically as well as biologically important, with applications ranging from catalysis, energy storage, and molecular electronics to medicine. Therefore, the achievements here will have a tremendous impact in X-ray chemical imaging of single molecules and the results will be potentially transformative.

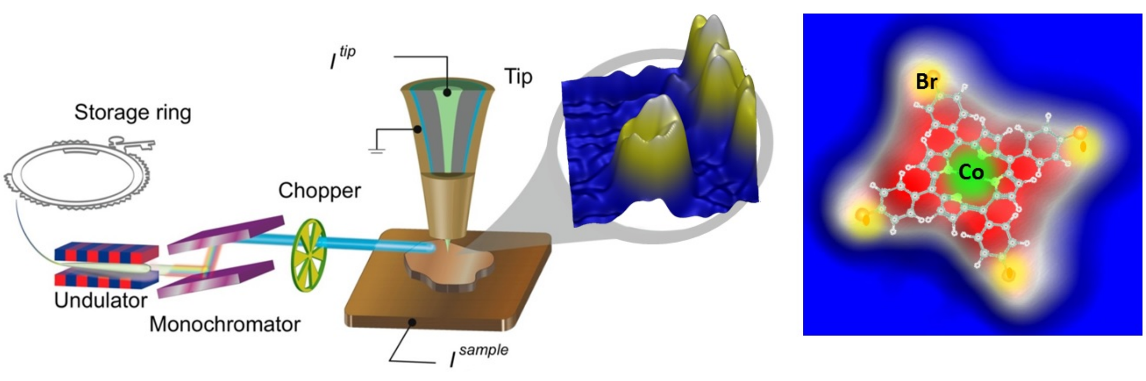


Figure 3-4. *In SX-STM, a synchrotron X-ray beam illuminating onto the tip-sample junction produces X-ray excited electrons that are tunneled to the tip and provides elemental contrast (left). An STM image of Co porphyrin molecule with chemical structure overlay for eye guidance (right).*

3.2.b. Theme II, Manipulating nanoscale interactions

A central motif here is to study and manipulate the forces, the interactions, and the energy dissipation between nanoscale constituents at interaction lengths that vary from distant (~10 nm), to the atomic scale. This interaction could be collective and at a longer length scale, such as the non-linear interaction on nano-electromechanical systems, collective but at atomic length scales, such as the prediction and discovery of materials using molecular dynamics, or individual and dissipative interaction at atomic length scales such as in friction.

Probing interactions at the atomic level: computational materials discovery

Materials discovery, and predicting microstructure evolution in materials from purely first principles physics using quantum mechanical interaction between collections of atoms is still computationally intractable. On the other hand, there is an opportunity to apply approaches developed in machine learning and data science, in concert with first principles physics to develop accurate yet efficient computational approaches to materials discovery and predicting microstructural dynamics and behavior. The TMG Group, also in collaboration with the EXM and QEM Groups will work to pioneer materials discovery combining first principles theory and experimental observations with data science techniques. We will explore new means of determining force fields for large-scale molecular dynamics (MD) simulations capable of describing reactive catalytic processes, disorder and phase transformation much more accurately and efficiently than is possible today. Applications are being and will be made to 2-D materials, nano/meso-scale phenomena (e.g. grain-formation, phase transformations) in bulk materials such as water, and low energy defect formation in materials for neuromorphic processing.

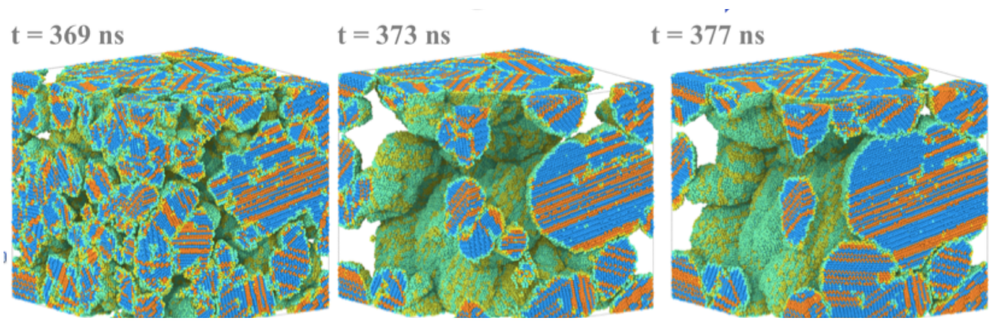


***Figure 3-5**. Large scale MD simulations of ice grain growth in water are enabled by both a highly-efficient machine-learning-based force field and a new, unsupervised learning techniques for identifying grains, resulting in better accuracy and ~100X improvement in computational efficiency.*

Dissipative interactions at the nanoscale: Friction and Superlubricity:

Fundamental understanding of the atomistic scale dynamical processes at tribo-interfaces are crucial for the design of functional lubricants. Recent experimental studies have shown that 2D materials (including MoS_2 and hexagonal-BN)) when combined with nanoscale diamond particles, undergo tribochemical reaction at the nanoscale leading to formation of onion-like carbon (OLC) structures at the tribological contact during the sliding process (Figure 3-6) resulting in superlubricity at macroscale. We will be carrying out detailed experimental and theoretical investigation in understanding the mechanism of tribochemical/catalytic reaction induced transformation of 2D materials leading to superlubricity. We will be exploring the materials phase space to identify other 2D materials (in combination with nanoparticles) that could lead to superlubricity with the aim to develop an overall materials genome that may enable designing lubricants. This work is a close collaboration between the TMG group (predictions/materials discovery) and the Nanofabrication Group, along with the Energy Systems Division at ANL.

Collective interactions at large (~10 nm) length scales: Metasurfaces and NEMS

All bodies are surrounded by fluctuating electromagnetic fields due to thermal and quantum fluctuations of the charge and current density at the surface of the bodies. Immediately outside the bodies, this electromagnetic field exists partly in the form of propagating electromagnetic waves and partly in the form of evanescent waves that decay exponentially with distance away from the body's surface. In this research theme, we intend to implement reliable methods to probe, control and manipulate nanostructures by controlling these near-field forces. By integrating nanoscale optical resonators with micro mechanical actuators we can precisely control the enhanced and highly localized electromagnetic fields afforded by coupled plasmons, without negatively affecting their optical properties due to their inherent extreme sensitivity to the local environment. We plan to use this scalable and reconfigurable optical nanocavity as a flexible platform for the development of chip-based plasmonic circuitry and hybrid nanophotonic systems, which demand precise control of the near-field coupling between plasmonic modes and a variety of nanostructures like single photon emitters, electron spins, and nanomechanical devices. Furthermore, by integrating arrays of optical nanoscale resonators with NEMS devices we intend to develop reconfigurable metasurfaces capable of delivering optical properties on demand. We have recently demonstrated dielectric metasurfaces that can manipulate light in the visible spectrum with almost perfect efficiency (see Figure 3-7). These metasurfaces will make it possible to create ultrathin optical components that control light in radically unique ways. By controlling the spatial distribution and dimensions of the dielectric resonators, it is possible to "design" the optical properties of a flat surface and thus, to "design" novel optical phenomena. Since the fabrication processes of metasurfaces are planar, these flat lenses could be easily incorporated into MEMS/NEMS devices and integrated circuits, making it possible to produce a novel generation of extremely thin optical systems. As the dimensions of mechanical devices are reduced to the micro and nanoscale, their

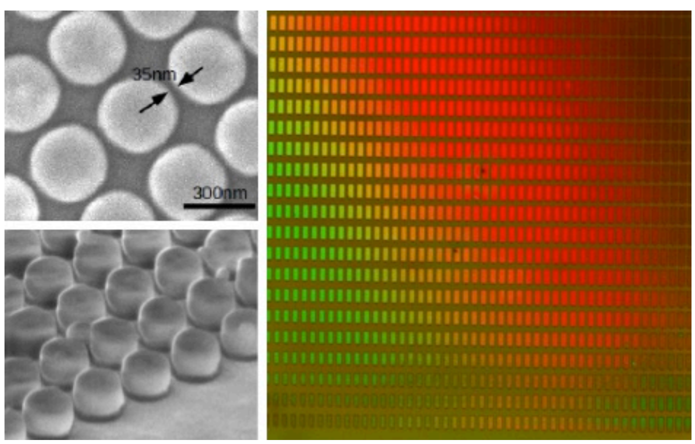


Figure 3-7. Left: Arrays of TiO_2 nanoparticles on glass. When such dielectric metasurfaces are illuminated with white light, the particles act as nanoantennas, allowing full transmission of predetermined wavelengths. Right: Arrays of nanoparticles having different gaps and radii under white light illumination.

dynamical behavior becomes strongly nonlinear. Moreover, due to the small size of these structures, thermal fluctuations become increasingly important and need to be taken into consideration when describing the physical state of the system. Here, we propose to study the complex nonlinear dynamics of micro and nanomechanical systems in the presence of classical fluctuations as a paradigm of fluctuating nonlinear mesoscopic systems. This research would allow identifying novel mechanisms to manipulate and control the state of nanomechanical systems capitalizing on the intrinsic nonlinear phenomena of micro and nanoscale resonators. These projects will be done collaboratively between the Nanofabrication and Devices, the nPBS, the QEM, and the Theory and Modeling groups.

3.2.c. Theme III, Hierarchical synthesis for energy, information and functionality

This theme aims to achieve energy efficiency and energy transduction in systems that utilize hierarchical synthesis and materials design at the nanoscale. It combines synthesis and nanofabrication across different scales and aims to use both self-assembly and top-down approaches. The objectives for this approach, in the end, are to identify new pathways in energy and information transduction including propagation of charge, spins and collective phenomena; and develop new ways for adaptive responses to the environment.

Biofunctional structures

Biology remains the model and inspiration for complex, hierarchical, functional materials architectures. Photosynthetic energy conversion, recognition, signal transduction and amplification, neural networking, navigation, and sensing are among most stimulating concepts. We study processes perfected in the natural systems to develop and utilize hybrid nanomaterials and architectures, not found in nature, but are guided by nature's principles. For this purpose, we design and assemble energy gradient architectures composed of peptides, nanoparticles and proteins that enable distance dependence of charge and energy transfer. We integrate peptide amphiphiles into robust nanoreactor architectures (capsules that act as simplified artificial cells) for light-induced charge separation functionality. Nanoreactor architectures of colloidosomes (selectively permeable nanoreactors composed of colloidal nanoparticles) are also employed to achieve signal transduction and sensing. We introduce of electrochemical signals to nanoreactors by introducing light active proteins (e.g. opsins) or ion gradients (ion channels) and probe their ability for hydrogen generation in the presence of mineralized photocatalysts. We also develop dynamic porous nano-bio structures. By combining peptide engineering and metal coordination, we generate metal peptide frameworks (MPF) that allow the peptide to serve as flexible linkers while the metal binding sites serve as a photosensitive hinge, thus creating tunable photoresponsive architectures. Investigation of these structures involves X-ray nano imaging, for elucidating chemical structure and colocalization measurements under stimuli (EXM Group), hyperspectral time resolved optical spectroscopy (nPBS) and droplet microfluidics for manipulating nanoreactor functioning (NFD Group). In situ studies using liquid holder TEM will deliver critical understandings of forces involved in the process of peptide driven mineralization and nanoparticle self-assembly.

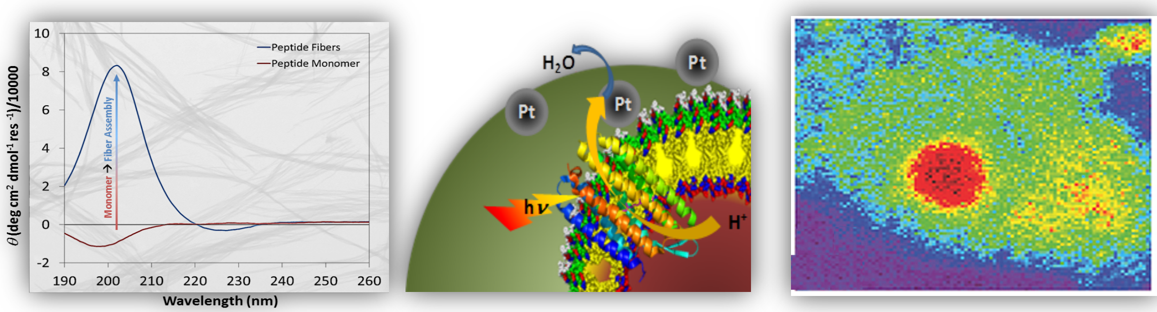


Figure 3-8. *Left: Design of self-assembly of peptides in different secondary structures used for assembling of artificial cell (Middle). Right: X-ray nano-imaging of colocalization of Pt upon interfacing engineered thermosensitive magnetomicelles with a cell.*

Neuromorphic materials

At the heart of revolutionizing computing is the quest for new neuromorphic, computational architectures for manipulating information efficiently. Biologically inspired, but in form and function much closer to the inorganic world than the effort in biofunctional materials described earlier, these architectures require new materials and devices whose properties can encode the functionality of neuromorphic components such as neurons, synapses, and interconnects. Our program, will develop materials and systems that will offer this neuromorphic functionality at extremely low power levels. We will focus on new materials paradigms for the systematic development of new neuromorphic components where materials properties can be modified (switched, written and read) using ultra low energy/voltage at ~few hundred mV. We will accomplish this by tailoring materials at the atomic scale and look beyond the use of electronic degrees of freedom to an integrated control of electron, phonon, ion and point defect transport, and correlated phenomena such as insulator-to-metal transitions. These will result in the design of hierarchical material structures where a neuromorphic functionality is encoded in the response of the structure. An example is shown in Figure 3-9 where the simple integrate-and-fire operation of a neuron, can be encoded in a simple metal-to-insulator element modulated via joule heating. These studies will be guided via detailed molecular dynamics based simulations of material response for such functionality. The goal, in this study, is to provide the underlying materials science and understanding of the nanomaterials and structures that will underpin neuromorphic information processing engines of the future.

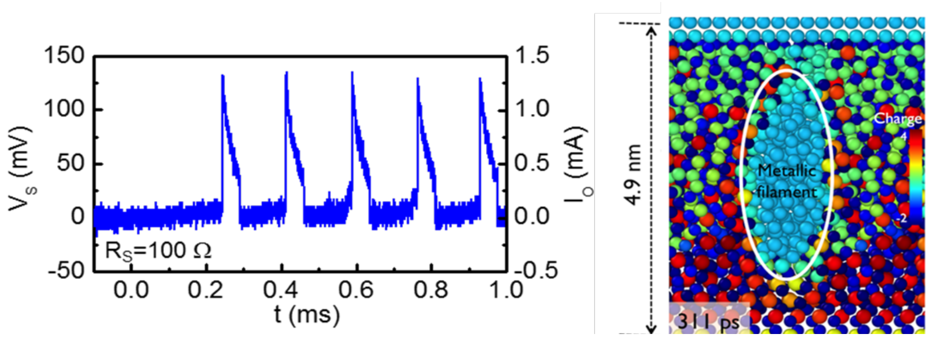


Figure 3-9. *Left: Experimental output waveforms of an artificial neuron when the input is a constant current clamp. The artificial neuron is based on a VO_2 insulator-to-metal,(IMT) transition element, Right: Formation of a metallic filament in a HfO_2-Cu synaptic heterostructure, after 311 ps of MD simulation, via electric field assisted growth.*

Hybrid nanostructures

Building upon promising results, our work here will focus on hybrid organic-inorganic nano-architectures with controlled porosities for selective filtration and absorption—areas with significant potential in the energy sector. In the sequential infiltration synthesis (SIS), invented at Argonne, we can uniquely enable self-limited growth of inorganic substances within polymer substrates using vapor-phase molecular precursors and controlling their diffusion and interactions with chemical moieties on the polymer chains. Even though only a small fraction of the phase space of possible precursor–functional group interactions have been explored to date, the work has resulted in the development of highly promising sponges for oil recovery (Figure 3-10). We will study new combinations of polymers and inorganic precursor species using in situ spectroscopy, quartz crystal microbalance, and density functional theory, in order to establish a a broad library of hybrid composite materials, enabling us to tailor affinity, reactivity, and other characteristics with unprecedented control and flexibility.

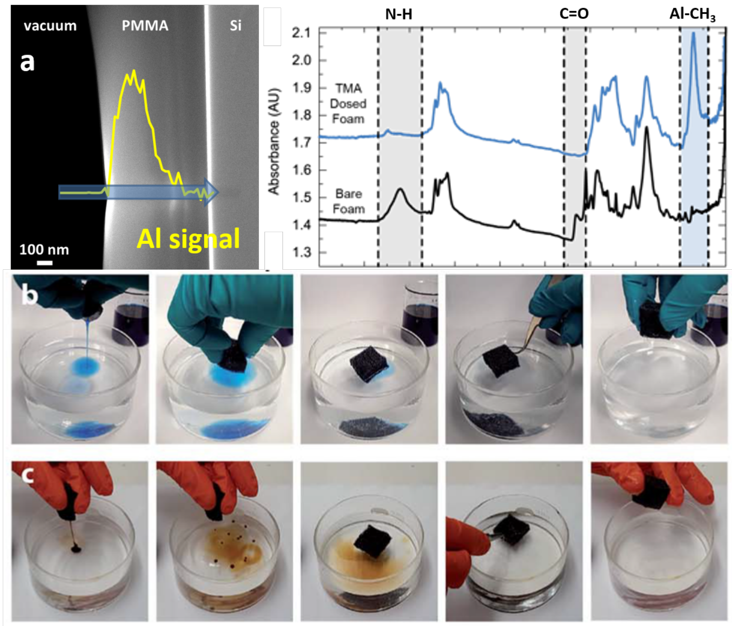


Figure 3-10. *a) (left) Cross-sectional SEM image of a thick poly(methyl methacrylate) film in which several hundred nanometers of aluminum oxide have been synthesized using SIS (overlaid with EDX signal) and (right) FTIR spectra for untreated polyurethane foam and SIS-treated polyurethane foam showing reactivity of the amine and carbonyl functional groups. b) Photographs of Oleo Sponge selectively adsorbing dyed silicone oil (blue) and crude oil (brown) from a water bath.*

Similarly, we are using a self-assembly process to create free-standing membranes made of nanoparticles, such as Au, Fe_3O_4 nanoparticles, coated with alkanethiol ligand. The mechanical strength of these membrane exceeds GPs ranges, though they are much thinner than traditional polymeric membranes that require high applied pressure and energy. Most remarkably, the pores in between the nanoparticles allows controlled transport of molecular dyes depending upon their sizes and charge state. Based upon these progress, we will design and synthesize membranes with nanometer thickness through controlling self-assembly of various inorganic nanoparticles/nanowires and surface molecular ligand functionalization.

2D materials—Borophene and Beyond

As pioneers in the experimental discovery of borophenes (the first synthetic 2D material and new boron allotrope), we are now rapidly expanding our understanding and development of low-dimensional borophenes towards synthesis on new substrates, heterostructures, exotic physical properties (quasi-particle interference, charge density waves (CDWs), and superconductivity), and isotopically pure borophenes of ^{10}B and ^{11}B. Our work is aimed at direct synthesis of lateral heterostructures with other 2D materials such as graphene, investigation of the anomalous pseudogap at low temperatures, and the search for superconductivity which has now been theoretically predicted. We are also exploring the development, synthesis, characterization and processing of isotopically pure borophenes comprised of either ^{10}B or ^{11}B, and the examination of ^{11}Borophene for neutron detectors.

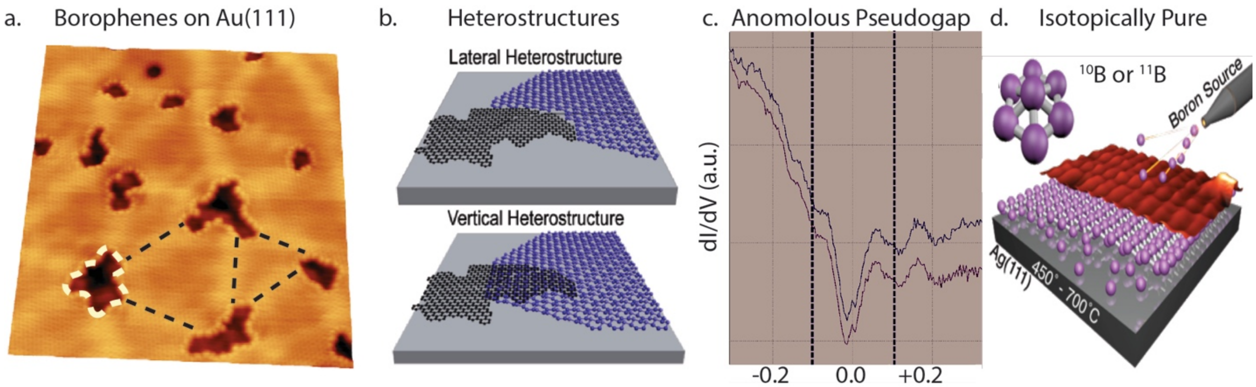


Figure 3-11*: A sampling of our high priority objectives to advance the field of synthetic 2D materials. (a) Initial nucleation of borophene on a Au substrate. (b) Borophenes interfaced with other 2D materials to form heterostructures. (c) Scanning tunneling spectroscopy data showing an anomalous pseudogap. (d) We will work to synthesize isotopically pure borophenes along with digital heterostructures.*

3.2.d. Vector capabilities in electron microscopy and the hard X-ray Nanoprobe supporting themes I thru III

All the studies performed in Theme (I) through (III) include requiring detailed atomic understanding of temporal and spatial structural response to applied stimuli. The imaging of strain is a central theme in our vision for our activities in electron microscopy and the hard X-ray Nanoprobe facilities. Two examples are in its role in tuning the energy states of qubits (Theme I) or controlling the interaction between them (Theme II). While a detailed strategy for electron microscopy and the hard X-ray Nanoprobe will be completed by April-May 2017, the following is a summary of our vision for these two areas.

Electron microscopy: (more detailed inputs forthcoming by Apr/May 2017). *Towards Smart, Dynamic, and Correlated Imaging – Leveraging Data Science*: The electron microscopy group at CNM has a unique expertise with a combination of techniques including chromatic aberration-corrected high-resolution transmission electron microscopy and in-situ characterization of materials behaviors. Building up these strengths, we propose dynamic and correlated Imaging that integrates data science and detector/holder development. The strategy aims to address key fundamental challenges in electron microscopy: effective detection and dynamic extraction of weak/indirect signals for studying structural, chemical, functional properties of quantum materials, nanoscale hybrid materials, and transformations of energy materials in operando conditions. The strategy consists of three scientific approaches: detecting weak/indirect signal (smart), rapid operando changes (dynamic), and complementary properties (correlated) (Figure 3-12). On the hardware side, we will develop and apply new pre- and post-specimen phase plates, platforms enabling control of external stimuli, and enable detection with high-speed high dynamic cameras. We will develop holders, multiple-probe and multiple-detector configurations, which will enable simultaneous use of multiple imaging and spectroscopic characterization methods. On the software side, we will develop and leverage algorithms and tools in data analysis, workflow, atomistic modeling, simulation, computer vision, and machine learning, which will be used iteratively to optimize imaging conditions and experimental conditions, as well as enable accelerated interpretation of imaging results, including from multiple imaging techniques.

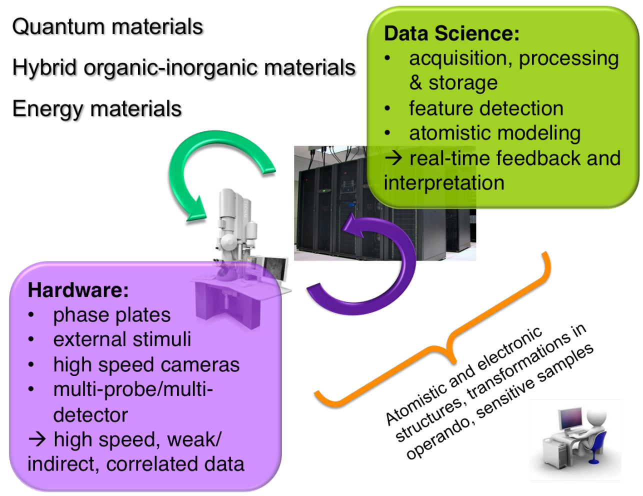


Figure 3-12. *Illustration of strategic directions for electron microscopy*

X-ray microscopy: (more detailed inputs forthcoming by Apr/May 2017).

The CNM/APS HXN harnesses the brilliance of the APS 3rd generation synchrotron source for transformative insight into nanoscale materials, in particular leveraging the best-in-class per-bunch source brightness for the study of stimulated dynamic nanostructured phenomena. Consequently it is a unique platform for observing dynamic effects in nanoscale materials research, enabling nondestructive volume probing of sub-pm strain displacements at 3D voxel resolutions of ~20 nm, and correlative imaging of strain with chemical speciation at parts-per-million trace elemental detection limits. It is also equipped with a stable and well integrated scanning temperature stage (cryogenic - 600K), a liquid / gas flow cell for in-situ chemical experiments, and electrical feedthroughs for device switching that enable in-situ, in-operando measurements and the study of phase transitions. The work is supported by advances in Bragg Projection Ptychography (BPP) analysis, made in collaboration with APS and MSD, that has enabled the demonstration of 5 nm resolution in 2D and the demonstration of fixed angle 3D ptychography. Both of these are substantial results in the field and were recently enhanced by the implementation of GPU-based reconstruction algorithms in collaboration with Mathematics and Computing Sciences Division at ANL, that increased our reconstruction speed by factor of thousand. Looking forward, the CNM/APS Hard X-ray Nanoprobe is targeting in-operando, dynamic measurement of strain in material volumes, taking advantage of the exceptional per bunch brightness of the synchrotron light source, with applications in a range of areas such as strain effects on defect dynamics, chemical catalysis and corrosion, and local critical phenomena in phase transitions. Following the APS-Upgrade (APS-U) anticipated in 2022 that will provide ~100x improvement in coherent beam intensity, HXN upgrades are planned that will increase the range of these dynamic studies - enabling video-frame-rate imaging of irreversible structural phenomena at ~10nm spatial and ~10ms time resolution, and 3D BPP visualization of cubic micron volumes at ~20nm spatial and ~100ps time resolution. This two orders of magnitude increase in focused flux will also make possible nano-focused X-ray Diffuse Scattering methods to observe phonon and plasmon dispersions used for nanoengineered heat transport.

3.2.e. Vector capabilities in computational materials science supporting themes I thru III

The exponential growth in computing power and the availability of highly scalable atomistic simulation codes are revolutionizing the computational analysis of materials. The ability to simulate larger volumes of a dynamically changing material via different computational techniques is traded off by the time resolution at which the dynamics is monitored (Figure 3-13). Concurrently, the advances in computing hardware have also revolutionized data analytics and machine learning or, more generally, data science. Going forward, these powerful techniques of data science can play a significant role in computational materials science by combining data analytics with first principles physics and computational physics in general. These developments have also led to the recent Office of Science and Technology Policy (OSTP) initiated thrust on materials genomics, so that we anticipate extensive user interest on this topic.

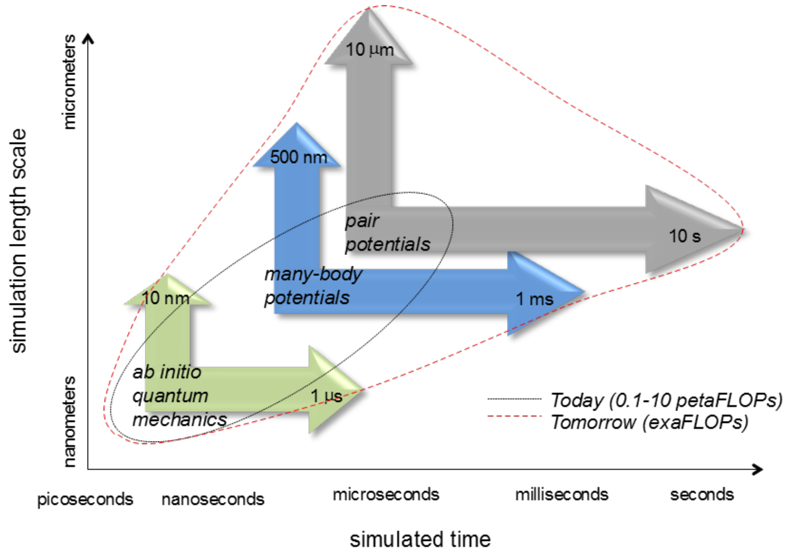


Figure 3-13. *The trade-off space between simulated volumes and time slice resolutions for simulations of dynamically changing materials. Capabilities today (with peta-FLOP computing) are compared against future exa-FLOP computing capabilities. (From A. Curioni, IBM).*

Our work will focus on the following areas in order to support themes I through III:

2. **Develop computational methods to describe quantum materials and phenomena**:
We will develop and apply approaches to simulate qubit interactions in hybrid nanostructures and other quantum phenomena, including the dynamical evolution of excitions. This work will involve both quantum master equation approaches allowing for environmental effects and first-principles calculations including density functional theory and many-body perturbation theory. The work will also employ recent developments in sparse-matrix representations and optimization techniques in order to efficiently and accurately simulate experimentally relevant systems such as defect states in WSe_2.
3. **Use of machine learning and first principles physics to develop user-accessible force fields for molecular dynamics**: Despite advances in electronic structure methodologies/implementations, there is still a substantial gap between time and length scales accessible to ab initio molecular dynamics vs more computationally efficient but less accurate semi-empirical functional forms based on classical force fields. To meet this challenge, there is an opportunity to develop a new class of inter-atomic potential functions or force fields that combine the accuracy and flexibility of electronic structure calculations with the speed of classical potentials by merging and exploiting the best insights from the fields of machine learning, advanced optimization and atomistic simulations. We will develop such force fields to describe electrochemical processes including nanoscale aqueous corrosion, 2-D and heterogeneous material properties, and grain formation and phase transitions in bulk materials (e.g., water).
4. **Developing machine learning techniques to connect multi-modal imaging to atomistic modeling:** We will use atomistic and first-principles modeling for X-ray, electron and scanning probe microscopies to produce simulated imaging data, which can be combined with experimental data and processed with pattern recognition, multi-objective evolutionary optimization, and other machine-learning approaches to develop a more complete and accurate nanoscale understanding of structures and transformations, and to accelerate the discovery of new functional materials.

4. Development of Synergies with other DOE User Facilities at Argonne

The CNM leverages its science and its capabilities with the strengths of the other BES user facilities co-located at Argonne National Laboratory. Examples of such synergies are provided below.

4.1 Hard X-ray Nanoprobe Facility

The CNM's Hard X-ray Nanoprobe facility, located at Sector 26 of the APS, was jointly built and operated by CNM and APS. It is the only dedicated X-ray microscopy facility within the portfolios of the nation's five Nanoscale Science Research Centers and its nanoscale imaging capabilities are best-in-class for experiments involving dynamic measurements of materials. The CNM directs the scientific program, manages the operations, and provides the majority of the funding for the Hard X-ray Nanoprobe. APS provides scientific and technical support to the Hard X-ray Nanoprobe through an in-kind effort contribution. Experimental beamtime at the Hard X-ray Nanoprobe is allocated on a 75% - 25% basis between CNM and APS. Proposals for beamtime may be submitted either through the APS or CNM proposal submission portals. As part of our collaboration with APS we are adapting the microscope infrastructure to deliver transformative microscopy of dynamic systems and thermal transport harnessing the diffraction limited storage ring of the APS Upgrade. Details of the technical upgrades planned for this are described in Section 6.

4.2 Dedicated Beamline for Synchrotron X-ray Scanning Tunneling Microscopy

A successful example of the CNM synergy with APS is the recent development of the world's first dedicated SX-STM beamline, called "XTIP", at the Advanced Photon Source. Currently, a new beamline is being built at APS Sector 4 to take full advantage of the brightness and polarization control of the undulator source there. XTIP will be completed in 2019; the SX-STM program will receive 20% of the beamtime at beamline 4-ID in the interim, during the XTIP construction period. We have now begun working with a limited number "beta-users" on the early version of the SX-STM, to begin to fully develop the capabilities of the technique. By 2019, with the completion of the construction, the SX-STM will be a fully accessible tool available to the community.

4.3 MEMS X-ray Pulse Selector

Recent collaboration between CNM and APS also led to the work that resulted in a *MEMS X-ray pulse selector*, a new class of devices for controlling subnanosecond timing of the delivery of X-rays. Shrinking of X-ray optics to the microscale using MEMS technology, created an opportunity for developing ultrafast devices that reflect X-rays at precise times and specific angles. Recent experiments demonstrate that these devices can achieve sub-ns gating windows (~500 ps), ~2 orders of magnitude better than the typically used mechanical choppers. This work is expected to lead to compact, sophisticated X-ray optical approaches for studying the structure and dynamics of matter at atomic length and ultrafast time scales (Figure 3-2) and lay the groundwork for the development of a suite of X-ray optics, e.g. ultrafast gating devices, multiplexers, ultrafast spectrometers/monochromaters, to facilitate experiments currently not possible at X-ray synchrotrons.

4.4 Integrated Computational Tools

The *Theory and Modeling Group* has strong associations with Argonne's Leadership Computing Facility (ALCF). For example, DOE INCITE projects entitled "Mesoscale reactive simulations of electrochemical interfaces" and "Combining high accuracy electronic structure methods to study surface reactions" are part of CNM's research portfolio. These projects involve significant amounts of computer time on some of the fastest supercomputers in the world and also involve working closely with ALCF staff members on developing and improving the performance of molecular dynamics codes (LAMMPS and NAMD) and on quantum Monte Carlo electronic structure approaches. Members of the ALCF also participate in the *Theory and Modeling Group's* group meetings.

4.5 Partnership with the Laboratory

Argonne National Laboratory is in the process of building an additional *6000 sq ft of cleanroom space* connected to the CNM's existing cleanroom (Figure 4-1). The extension of the existing cleanroom will house prototyping and testing equipment to enable the advancement of nanoscale devices from the research phase to development. Construction is planned to end June 2017. The cleanroom extension and its capabilities, while not a part of CNM, will be available to CNM users.

***Figure 4-1**. The Argonne Cleanroom under construction*

Argonne is also in the process of constructing a *liquid helium recovery system* in order to recover helium gas currently being lost from the boil off and helium transfers of cryogenic instruments. The CNM will have a recovery & compression facility that will transport captured helium to the liquefaction plant located in the Physics division, the largest consumer of helium on site. After liquefaction helium will be delivered back to CNM for recycled use. Installation of the helium recovery facility is expected in 2017.

5. Crosscutting Research with other Programs

5.1 Crosscutting Research with Core Research Programs at Argonne

Science and capabilities at the CNM leverage core research strengths with several research programs at Argonne:

Computational materials: The CNM Theory & Modeling group is a participant in the Center for Electrochemical Energy Science (CEES, a DOE Energy Frontier Research Center), the Midwest Integrated Center for Computational Materials (MICCOM, a DOE-Basic Energy Science funded Center), SunShot Bridging Research Interactions Through Collaborative Development Grants in Energy (BRIDGE, a DOE-Energy Efficiency and Renewable Energy program), and a Strategic Partnership Project (SPP) with Toyota Research Institute of North America (TRINA). In CEES, we have developed computational approaches to configurational sampling for non-equilibrium electrochemical processes in high capacity lithium-ion and beyond-lithium-ion energy storage materials, and a computational capability for modeling Raman spectra. In MICCOM, we are developing scale-bridging capabilities for the modeling of solid-liquid interfaces and thermal transport in nanostructured materials. In the BRIDGE project, we are developing a high throughput computational framework for sampling and evaluating grain boundaries in semiconductors. As part of the SPP with TRINA, we have developed methodology to study the electronic and thermal transport properties of metal-insulator materials. These new capabilities strengthen and expand the intellectual and scientific expertise that is available at the CNM.

Photon Dynamics: Our expertise in time resolved spectroscopy has contributed to a DOE BES ultrafast science project that aims to determine impacts of coherent phenomena on electronic processes such as molecular motion-driven photocatalyst activation. In this project the CNM is making time-resolved optical measurements of nanomaterials that exhibit coherent vibrational motion and correlating them with electronic phenomena such as energy transfer. The results aim to connect to nonequilibrium energy flow and to photocatalysis,

Quantum Materials: The CNM is collaborating with a DOE BES Materials Sciences Program with the Institute for Molecular Engineering (IME, a partnership between the University of Chicago and Argonne) entitled "Quantum Metamaterials". The intent of the collaboration is to establish a vibrant quantum materials and sensing research effort within Argonne, and to benefit from the deep expertise that currently exists at IME on the subject. The goal is to explore the physics of quantum states and entangled states for energy efficient information processing.

Synthesis: Sequential infiltration synthesis (SIS), a novel materials synthesis technique invented and developed at CNM in partnership with researchers in the Energy Systems Division at Argonne, has generated interest in diverse research communities. Within Argonne, SIS has been leveraged in programs supported by the U.S. Coast Guard for oil spill remediation, JCESR for novel materials for energy storage, and the IME for functionalization of polymeric materials. Moreover, a budding program at Argonne built on materials science and engineering for water research will fold SIS techniques into many topical studies for fluid separations and membrane science. Beyond Argonne, SIS has been adopted in active research at IMEC for next-generation lithography technology, at other NSRCs such as CFN, and in other industrial research laboratories.

5.2 Partnership with Users

Several partner user proposals (PUP) are in place at this time with various Argonne divisions:

THz physics: the CNM is developing time-resolved terahertz (THz) spectroscopy that leverages a femtosecond amplifier in the CNM laser labs, while the APS funds the construction of a THz spectrometer. THz spectroscopy is valuable for studying a large range of condensed matter phenomena, including, for example, phase transitions in complex oxide nanomaterials and coupled spin and valley dynamics in two dimensional transition metal dichalcogenides. When complete, CNM staff and users will gain a new, cutting-edge capability for characterizing nanomaterials at THz frequencies including THz time-domain spectroscopy, THz-pump THz-probe, THz-pump optical-probe, and optical-pump THz-probe. These capabilities will complement newly developed THz pump X-ray probe capability at the APS.

Superconducting bolometers for the South Pole telescope: A PUP with Argonne's High Energy Physics division involves a research and development effort with the CNM to fabricate large arrays of multi-chroic Transition Edge Sensor (TES) bolometers for use in Cosmic Microwave Background (CMB) experiments (Figure 5-1). The focus is on developing techniques to implement and control the lateral proximity effect and to reduce two-level-system loss in superconducting microstrip at mm-wave frequencies. The goal is the stable and robust production of multi-chroic microstrip-coupled TES bolometer arrays across multiple 150 mm substrates. Several tools installed in the CNM clean room as part of this PUP are available for users (an ASML stepper, an etcher, and two AJA deposition tools).

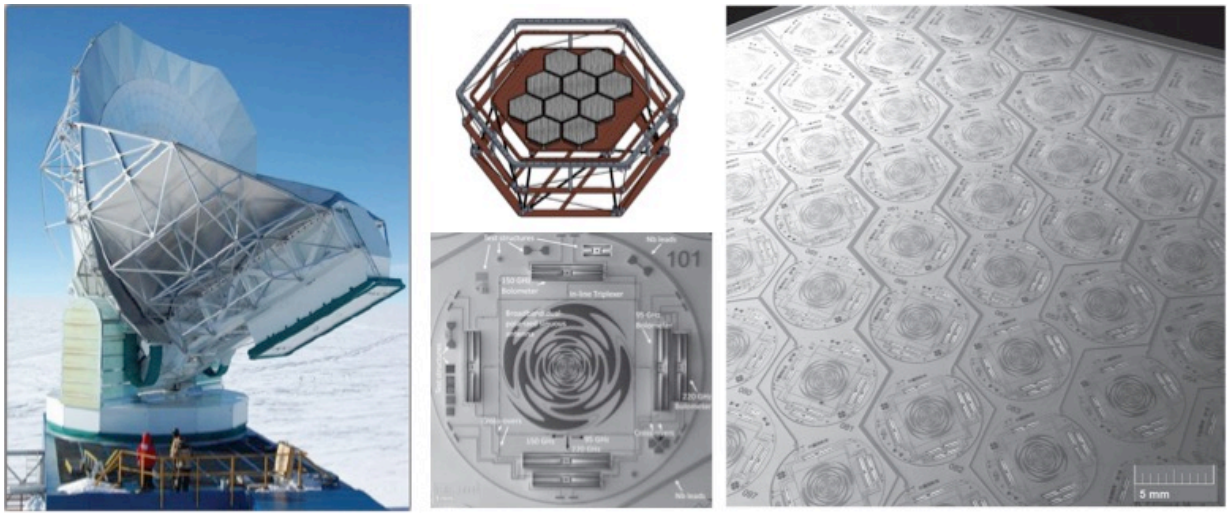


Figure 5-1. *Clockwise from left: South Pole Telescope where the next-generation CMB polarization experiments are performed; CAD drawing of the SPT-3G focal plane and support structure; SEM micrograph of a fabricated SPT-3G multichroic pixel array, including the wiring layout; and SPT-3G multichroic individual pixel.*

Finite Element Simulations: This PUP with APS utilizes the HPC Carbon cluster for finite element simulation. The PUP allows APS access to the CNM licensed COMSOL finite element software for development of capabilities that can help APS to design better scientific instruments. APS enhances COMSOL capabilities by adding modules that allow APS and the CNM user community to use multiphysics simulation capabilities. Some projects include nanopositioning for X-ray optics and samples, design of an acoustic levitator to dispense nanoliter samples, ultra-high heat flux front-end and beamline components, design of mirror bending and focusing using piezo-electric actuators with nanoscale actuation, and design of a nano-size beam analyzer.

Polarizers and coherent diffractive imaging: A partner user proposal with APS encompasses hard X-ray polarizers and dichroic coherent diffractive imaging for development of a new capability for imaging nanomagnetic stucture simultaneously with crystal lattice strains. This project advances our goals to develop dichroic coherent imaging capabilities at the CNM Hard X-ray Nanoprobe and SX-STM programs at beamline 26-ID, and to position the Nanoprobe to take full advantage of the unprecedented 100-fold increase in coherent flux to be provided by the APS Upgrade.

5.3 Industrial Outreach

One of the goals of the CNM is to increase industrial user participation in order to more fully embrace nanotechnology aspects and relevance to applied technologies. Key long-term industrial partners have included researchers from large companies such as Corning, Inc., Toyota Research Institute of North America, IBM, HP, GE Global Research Center, and small businesses such as Creatv Microtech, Advanced Diamond Technology, and OptoNet, Inc. Awareness of the CNM's user capabilities within the industrial community has expanded recently to include Toyota Motor Engineering & Manufacturing North America, Inc., Brewer Science Inc., several small businesses, as well as the integration of industrial users such as BP and UOP LLC; the latter are large oil companies primarily interested in catalyst development when accessing CNM. The CNM's industrial users have accessed all aspects of our capabilities from the HXN beamline to the nanofabrication facilities, and from various microscopy techniques to the high-performance computing cluster. The CNM maintains an "Information for Industrial Users" page on the CNM public website which includes industrial research highlights as examples for industrial scientists to benchmark their needs to what CNM can offer (http://www.anl.gov/cnm/user-information/industrial-users).

6. Key Instrumentation Decisions

Strategic approach

Equipment investment and recapitalization is important for the vitality of the CNM user facility and to successfully carrying out our thematic research. With this in mind, the CNM management team considered the following objectives when identifying and prioritizing planned equipment procurements:

- Unique capabilities that will attract high impact users and non-regional users
- Equipment that would advance our research themes.
- Replenishment, considering that equipment placed in the CNM is now approximately 10 years old.

Input for creating the CNM equipment plan came from the user community and CNM staff, through formal (equipment proposals, focused workshops, CNM Users' meeting, CNM strategy retreat), and via informal discussions.

With this approach in mind, our equipment plan is organized below by research theme with a 5 year time horizon, assuming that 10% of operating funds will be invested in equipment annually. This is not a comprehensive list, but a list of key items.

Quantum materials and phenomena

The CNM planned (and FY17 installed) equipment procurements in support of this theme are designed to advance directions in quantum optics, quantum sensing, quantum materials discovery, and modeling:

- Single photon detector, correlation function determination: In FY17 we developed a new CNM capability (available to users) to perform single photon detection in the near-infrared spectral region. This capability uses two superconducting nanowire single photon detectors (SNSPDs) to enable determination of the correlation function between single photons emitted from two coupled sources.

- In FY17 we will procure the primary components of a magneto-optical photoluminescence microscope cryostat tool that will also support optically detected magnetic resonance. The system will be capable of photon correlation studies of single particles emission and will enable spin coherence and control studies. Completion of the 9 Tesla instrument is scheduled for FY18.

- An upgrade of our scanning tunneling microscopy capabilities to include a q+AFM module is being initiated in FY17. The relative ease of use of the upgraded STM will enable greater user throughput in support of the quantum materials and phenomena theme (available to users in FY18).

- The CNM computing cluster: In FY17 we are procuring an Infiniband switch and 10 cluster nodes. Over the course of the next five years we plan to continue to replace older computer nodes and storage with current hardware, thereby increasing our computing capacity while lowering power consumption.

- We propose development of a mK temperature (less than 50 mK) scanning tunneling microscope coupled with an internal magnetic field strength of 9T or higher within the next 5 years. This system will also be the lowest temperature STM available for users among the NSRCs.

- We plan the development of time-resolved combined capabilities of transient photoelectron spectroscopy (trPES), time-resolved electron diffraction (trED), and time-resolved cathodoluminescence (trCL) in the next 5 years to enable direct monitoring of the temporal and structural evolution of excitonic interactions and charge separation in hybrid nanomaterials.

- We are currently building the world first beamline dedicated for SX-STM, “XTIP”, at Sector 4 of the Advanced Photon Source. This new undulator beamline offers circularly polarized X-rays and an energy resolving power of 4000 over the 450-1600 eV soft X-ray range.

Manipulating nanoscale interactions

A significant consideration in our equipment plan is to support the key nanofabrication capabilities. This includes the replacement or upgrade of equipment that was installed 10 or more years ago.

- In FY17 we are procuring an inverted microscope for staff and user science work related to planar optics, NEMS, and metasurface studies.

- In FY17 we are procuring a tribometer with Raman spectroscopy capabilities in order to support very successful research directions in low friction graphene and ultrananocrystalline diamond materials. This is a unique opportunity for users of the CNM.

- Within the next 5 years, we plan to procure a new e-beam lithography system (such as a JEOL JBX9500) to replace e-beam lithography capabilities installed more than 10 years ago in the CNM.

Synthesis of nano-architectures for energy, information and functionality

The equipment procurement plans for this theme relate primarily to the characterization of collective interactions of assembled or fabricated nanostructures for information processing and energy transduction. Procurements plans include:

- In FY17 we are procuring electrical test equipment designed to characterize structures for neuromorphic materials.
- In FY17 we are procuring droplet sorter equipment, based on an inverted microscope with nano- and microfluidics capabilities, designed to characterize biofunctional nanomaterials for energy transduction in both synthetic cell membranes and natural cell environments.
- An optical parametric amplifier (OPA) with a narrowed spectral width and a picosecond (ps) pulse was recently installed. The ps-OPA offers tunability from 470-2400nm and can be implemented in a variety of experiments.

Investments in the Hard X-ray Nanoprobe

Finally, in support of all of the themes, we are investing in collaborative efforts with the Advanced Photon Source to ensure that our strategic directions for development of the HXN at the Sector 26 beamline will make full use of the MBA lattice envisioned for the APS Upgrade, and the two-orders-of-magnitude greater brightness it will deliver. The impact of 100x phase-coherent nano-focused flux will not simply make our current microscopy methods better, but will be fundamentally transformative in two key areas: i) time domain ptychography, and ii) inelastic and weak-scattering microscopy. We plan a strategic investment in both these areas resulting in new HXN capabilities: i) time resolved video frame rate (~10ms) and pump/probe (~100ps) ptychographic strain imaging, and ii) nano-resolved studies of lattice excitations along with spin, charge, and orbital ordering in systems such as complex oxides. This investment will bridge the gap between currently "possible" and "impossible" microscopy experiments fully capturing the two orders of magnitude coherent flux increase in order to enable transformative science of dynamic systems with the CNM/APS HXN instrument.

Instrumentation developments that we plan to pursue in this collaboration at the CNM include:

- In FY17, new Nanoprobe data acquisition software is being procured that will enable a superior user science interface and enhanced throughput of experiments at the HXN.
- In FY 17 Liquid / gas flow cell was developed for chemical mapping (importantly compatible with the JEOL 2100F TEM) and integrated with simultaneous laser stimulation by the creation of an optical pathway and laser control area for 26-ID-C (facilitated in part by APS) allowing in situ photocatalytic oxidation behavior of individual nanoparticles to be directly observed.
- Within the next 5 years high-speed high-range Nanoprobe scanning capabilities optimized for fast-frame-rate ptychographic Bragg diffraction microscopy imaging at the nanoscale will be purchased.
- Time-resolved optical excitation and microscopy of the sample surface using a long-working-distance objective and excitation laser will be acquired for optical studies. The photoluminescence detection and optical microscopy will be compatible with simultaneous nanoscale Bragg diffraction.
- The HXN strategic directions depend on fully delivered coherent flux from the Nanoprobe Beamline to the HXN instrument, to ensure this we will rebuild the M1 mirror system interior components to optimize figure error, coherent flux transfer and mechanical stability.

Investments in Electron Microscopy

In FY 16 we have purchased a new FEI Talos TEM/STM with analytical capabilities (EDC) and tomography. The advance spectroscopy during in-situ characterization will open several new research avenues at the CNM. Discussions for additional upgrades and planned purchases are ongoing, and so far include (this list will be further refined):

- In FY17 we are procuring a replacement of an energy dispersive X-ray spectrometer (a 21 year old capability) for improved discrimination in elemental analysis and also providing much higher data acquisition rates that will enable better mapping capability and improve overall efficiency.
- Detection and extraction of weak/indirect signals: a digital STEM with beam precession electron diffraction analytical experimental framework that offers a suite of beam precession imaging and advanced analytical experiments.
- Correlated microscopy using multiple beams and detector: Atmospheric Gas TEM Holder for JEOL HR TEM with custom Optical Fiber.
- Computational algorithms, data acquisition and storage for rapid data interpretation and implementing computer vision algorithms and software.

7. User Program and Outreach Activities

Proposals are submitted via an electronic form during one of CNM's three annual open calls. The research description is captured by standard questions in 1-2 pages and selections are made from CNM's various instruments and capabilities. After internal screens for safety and technical feasibility, proposals are reviewed by at least three members of the CNM Proposal Evaluation Board (PEB) for scientific merit. PEB scores and comments are ranked and allocated primarily by score until time is expended. Facility-wide, approximately 70% of submitted proposals are allocated; however, this is capability-dependent as some instruments are in higher demand than others.

The CNM user program continually strives to attract the highest-impact users possible, including researchers from across the country and around the globe. Figure 6-1 displays the diversity of CNM users as a function of their affiliation, showing nearly half of the 500+ users per year are from U.S. academia, with correspondingly lower percentages of non-Argonne users from international, industrial, and other government organizations. Figure 6-2 displays the diverse fields of research represented by CNM users.

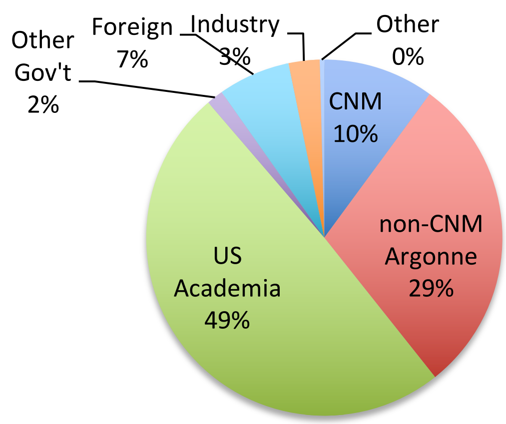


Figure 6-1. *Institutional affiliations of CNM users by affiliation during FY13-16.*

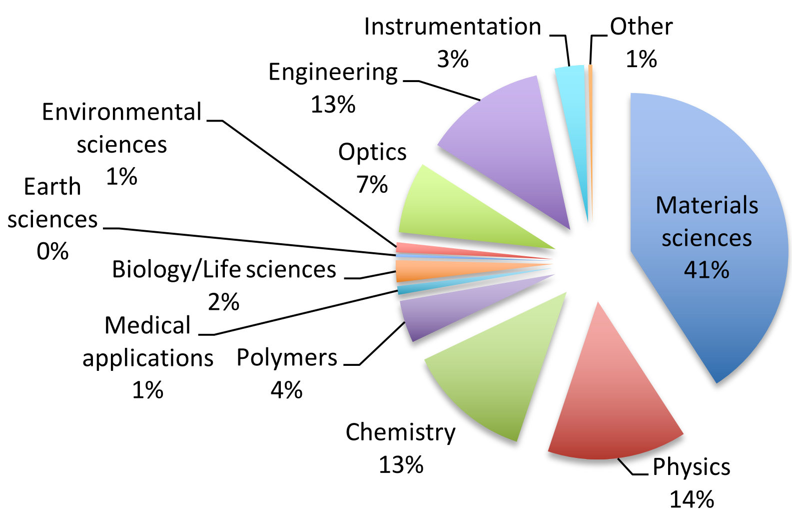


Figure 6-2. *Fields of research identified by CNM users during FY13-16.*

CNM's user partners are primarily a mix of university and government laboratory researchers—one of our goals going forward is to increase the participation of industrial users. To this end we are coupled to Argonne's NanoDesignWorks, which focuses on technology commercialization including the nanotechnology space, and to Argonne's Chain Reaction Innovation awardees. We also recognize a significant proportion of the CNM user community is affiliated with Argonne or research institutions within Illinois and could benefit from an infusion of geographical diversity. In order to expand the user base to capture additional national and international researchers in high-impact areas who could derive value from our resources, we will pursue several strategies such as:

- An updated communications and messaging approach will include a website reconfiguration for enhanced awareness, ease of use, and optimum analytics; CNM's unique capabilities will be featured prominently.
- Selected institutions are being targeted strategically for outreach visits by CNM staff.
- Figure 6-2 shows opportunities for growth in the numbers of CNM users in the areas of environmental, earth, biological, and life sciences. The nearby APS has large communities in these areas and so we are beginning to promote CNM to them with a new CNM column in the *APS User Newsletter*, information on the APS webpage, and by drafting CNM staff as associate members of relevant APS proposal review panels.
- Participation in joint NSRC outreach events such as promotional booths at AVS, APS, and TechConnect meetings.

The number of refereed journal articles is one of the primary quantitative metrics of project success at the CNM. The number of such journal articles authored by CNM users and staff combined in FY2016 was 293. CNM also dramatically increased the number of publications in the top echelon of journals – *Science* and *Nature* – from 2 in 2013-2014 to 8 in 2015-2016. The DOE recognizes a selection of 20 journals as conveying especially high-impact nanoscience and nanotechnology studies from the NSRCs to the public domain. In FY16, 26% of all CNM journal articles appeared on this NSRC high-impact journal list. We have set a target to increase the % of CNM publications in the DOE top 20 journal list to 33% (one-third), and to do so we have adopted the following strategies:

- Promote the top 20 list on the *For Users* tab of the CNM website (available here).
- Encourage CNM staff to publish in these journals when appropriate.
- Adopt and focus on the new scientific strategies as described elsewhere in this Strategic Plan, which are designed to target emerging high-impact nanoscience and nanotechnology areas.
- Target well-established and high-profile scientists as new users.

Other outreach activities initiated by the CNM are focused towards enhancing our existing user community and to growing the user base in areas of the highest scientific impact. An annual users meeting is held in conjunction with the APS during the second week of May to promote and enhance the latest research results within these co-located and complementary user communities. Promotion of our latest staff science results is accomplished via professional scientific meetings, invited institutional talks, press releases, our public website and social media, as well as hosting other scientific workshops throughout the year. Examples of these events are found on the CNM website at http://www.anl.gov/cnm/news.

8. Safety & Quality

The CNM has responsibility for environment, safety, health, and quality assurance (ESHQ) aspects of the facility's operations and, through policies and procedures, defines how responsibilities are delegated from the director through line managers to technically competent staff members supporting user research activities. The CNM program complements Argonne's laboratory-level safety program by incorporating methods, controls, and a work authorization approach tailored to the risk characteristics of a user facility and to the materials, instruments, and processes that constitute CNM operations. The specifics of the program will evolve in response to changing expectations, Argonne safety program evolution, and emerging information on hazards. Certified safety professionals help the CNM to better ensure research productivity and to ensure the program efficiently implements applicable ESHQ standards and requirements.

Furthermore, the CNM continues to employ a precautionary approach where there is uncertainty about hazard potential of new chemicals, including nanomaterials. This concept guides the conduct of hazards analysis and specification of precautions when handling nanomaterials. The CNM has continued its efforts to contribute to a better understanding and management of ESH concerns associated with nanomaterials and ESH questions for nano-enabled products. Examples of relevant activities include the following:

- The CNM continues to participate in Argonne efforts to promote a consistent and effective approach to dealing with ESH concerns about nanomaterials, primarily through Argonne's Nanomaterial Safety Committee.
- The CNM participated in the recent review of DOE Order 456.1, *The Safe Handling of Unbound Engineered Nanoparticles*, and we ensure the "best current knowledge is reflected in the identification and control of these potential hazards and impacts at their facilities."

Summary

In summary, we believe that the innovative science performed by CNM scientists shapes the user program while at the same time, innovative user science drives future scientific directions and capability development for the CNM. Sustaining this user-staff interaction, ensuring that the user base is distributed and diverse, making sure we remain relevant to user research needs in the future, and continuing to steward and shape the direction of the nanosciences remain our most important goals going forward.